Magnetoresistive Co/Cu multilayers: Hysteresis, polycrystallinity, and irreversible changes on magnetization

H. Holloway[a] and D. J. Kubinski[b]
Ford Research Laboratory, SRL/MD3028, P.O. Box 2053, Dearborn, Michigan 48121-2053

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Magnetoresistive metal multilayers are known to undergo an irreversible decrease in the maximum value of their field-dependent resistance after exposure to a magnetic field. An explanation for this effect in terms of the creation of antiphase domain boundaries is considered and rejected on the basis of experimental tests that point instead to a strong correlation of irreversible loss of resistance with magnetoresistive hysteresis. The main features of the phenomenon are reproduced by a model that treats polycrystalline multilayers as assemblies of grains with random orientations of magnetic easy axes. The key feature of the model is that for each individual grain the magnetic state corresponds to an energy minimum that may be only local, rather than global. Trapping in local energy minima accounts for both magnetoresistive hysteresis and incomplete antiferromagnetic order at the maximum resistance that is attained after cycling. The model explains experimental observations, such as the relatively small hysteresis and resistance loss at the first antiferromagnetic maximum. It also predicts a strong Co-thickness dependence of the resistance loss in Co/Cu multilayers at the second antiferromagnetic maximum that is verified with experimental data for Co/Cu multilayers. © *1998 American Institute of Physics.* [S0021-8979(98)03105-3]

I. INTRODUCTION

Multilayers (MLs) that alternate ferromagnetic and nonferromagnetic metals have been widely studied for their giant magnetoresistive properties. One of these properties that has been poorly understood is an irreversible decrease in the maximum value of the field-dependent resistance that is observed after an as-grown specimen has been subjected to a magnetic field that is sufficient to cause saturation.[1–4] This phenomenon has been observed with both the current-perpendicular-to-plane geometry and the more common current-in-plane geometry. (All of the measurements that we describe later used the latter geometry.) The effect is significant at the second antiferromagnetic maximum (AFM) of the Co/Cu system, but is apparently much smaller at the first AFM (Ref. 2, Fig. 3). It has also been seen in uncoupled Co/Ag MLs.

We first consider a possible explanation in terms of antiphase domain boundaries (ADBs), whose occurrence has been postulated previously,[5] and then describe some experiments that make implausible such a model for a decrease in the maximum resistance. Other preliminary experiments indicate a strong correlation between irreversible resistance loss and magnetoresistive hysteresis. From here we go on to consider the nature of magnetoresistive hysteresis in antiferromagnetically (AF)-coupled MLs. The approach that we take is related to that described by Folkerts[6] and Folkerts and Purcell[7] for algebraic treatments of specific orientations of the magnetic easy axis in single-crystal coupled bilayers, but here we use numerical methods to treat a polycrystalline assembly of ML grains that have randomly oriented easy axes. We find that the model accounts for a failure to attain perfect AF order, and hence the maximum possible resistance, after exposure to a magnetic field. Our calculations and experimental data relate specifically to Co/Cu MLs, but our model has general applicability to giant magnetoresistive MLs.

After describing our model for irreversible loss of maximum resistance, we consider its implications for the properties of magnetoresistive MLs. In particular, we compare its predictions with experimental results for Co/Cu MLs with varying Co thicknesses at the first, second, and higher AFMs (i.e., with Cu separator thicknesses of approximately 9, 20, 30,... Å).

II. SOME PRELIMINARY EXPERIMENTS TO TEST AN ADB MODEL

Figure 1(a) is a schematic resistance/magnetic field (*R/H*) plot that illustrates the concept of an earlier attempt at explanation that we based on ADBs. The as-grown ML is postulated to have perfect AF order at point A, as shown in the inset. With application of a magnetic field that is sufficient to give saturation, the ML reaches point B where it has complete ferromagnetic (F) order. With subsequent decrease of the applied magnetic field back to zero, nucleation of AF order can occur independently at different depths in the ML, as shown at point C. This can occur with ''mistakes'' in the phases of the AF-ordered regions so that their eventual coalescence gives rise to ADBs that persist at zero applied field, as shown at D. Thus, the resistance that is attained at point D after magnetic cycling is lower than the as-grown value at point A because the AF order is incomplete.

In this initial approach to the problem of understanding the irreversible decrease in maximum resistance after mag-

[a]Electronic mail: hhollowa@ford.com
[b]Electronic mail: dknbinsk@ford.com

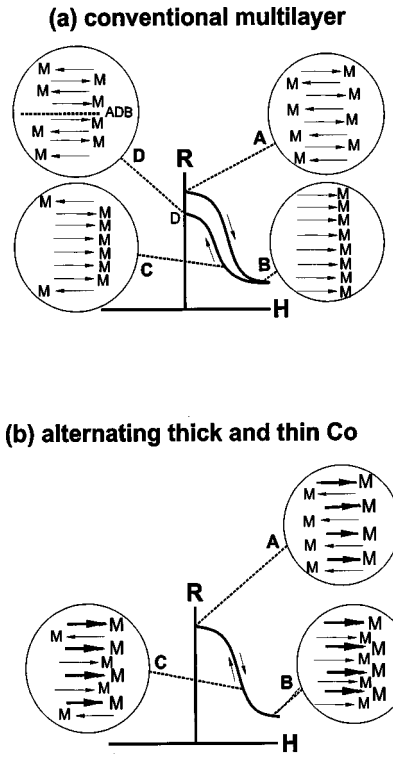

FIG. 1. Schematic illustration of the possible influence of antiphase domain boundaries. (a) A conventional Co/Cu multilayer. (b) A multilayer with alternating thick and thin Co layers.

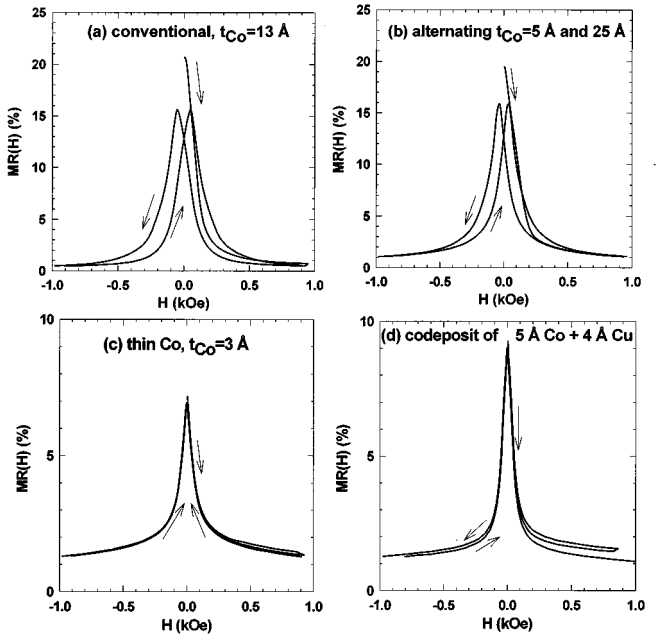

FIG. 2. Magnetoresistances at 300 K of specimens that were prepared to test the antiphase domain boundary hypothesis. (a) Conventional, $[Co(13\ Å)/Cu(20\ Å)]_{20}$. (b) Alternating thicker and thinner Co, $[Co(25\ Å)/Cu(20\ Å/Co(5\ Å)/Cu20)]_{10}$. (c) Very thin Co, $[Co(3\ Å)/Cu(20\ Å)]_{20}$. (d) Codeposit of Co and Cu, $[Co(5\ Å)+Cu(4\ Å)/Cu(20\ Å)]_{20}$.

netic cycling we were attracted by an idea that seemed promising for avoiding this change and, thereby, also avoiding the resultant loss of magnetoresistance after magnetic cycling. The complete failure of this approach was instructive in casting substantial doubt on the validity of the ADB model and in redirecting our search for an alternative explanation. For these reasons, we give a brief account of the preliminary experiments before continuing to a description of our new and more successful model.

In a previous article,[8] we described the properties of Co/Cu MLs in which consecutive Co layers alternate between two thicknesses. An essential feature of such MLs is that their thicker layers remain pinned by the applied field after its magnitude has been reduced sufficiently for the magnetization direction of their thinner layers to be rotated by the AF coupling to point in the opposite direction. In the context of the present problem, we reasoned that, even if nucleation of AF order were to occur independently at different heights in the ML stack, the pinning of the magnetizations of the thicker layers in the field direction would fix the phase of the AF domains, as shown at C of Fig. 1(b), so that their subsequent coalescence would occur without ADB formation, whence we should eventually return to the fully AF-ordered state at A of Fig. 1(b).

Figure 2(a) shows the resistance/field (*R/H*) characteristic of a conventional Co/Cu ML compared with that of an ML with alternating thicker and thinner Co layers shown in Fig. 2(b). (For the present work both the thicker and the thinner Co layers had thicknesses in the conventional range, unlike our previous study where the thinner layers were made exceptionally thin to reduce their hysteresis.) The observed decrease in maximum resistance after cycling in the two cases is quite similar: It is evident that our attempt to inhibit ADB formation has had no appreciable influence on the loss of resistance after cycling.

The preceding comparison seems to demolish the attempt to explain irreversible resistance loss as a consequence of ADB formation. Two further results suggested an alternative approach. First, Fig. 2(c) shows the results of cycling a specimen with very thin Co layers. We have previously described[9] the low-hysteresis properties of such MLs. Here the decrease in resistance that occurs on cycling is greatly reduced from that of the conventional specimen in Fig. 2(a). A similar reduction was observed with an ML which had the Co layers replaced by codeposited Co–Cu [Fig. 2(d)] where again there is reduced magnetoresistive hysteresis.[10] This correlation between magnetoresistive hysteresis and the reduction of magnetoresistance after magnetic cycling prompted us to seek a common origin for these phenomena. This is described in the following sections.

III. A MODEL FOR RESISTANCE LOSS

We start by considering the energy of an infinite stack of Co/Cu bilayers that have a single crystallographic orientation and a common easy axis that lies in the film plane. The direction of an applied magnetic field is taken to be the x axis and it is assumed that the magnetization directions of the Co layers alternate between two values, θ_1 and θ_2, relative to this axis. (This condition is relaxed later, when we

consider a finite stack of bilayers.) The energy per unit area of a Co/Cu/Co/Cu unit in an applied field H is then

$$E = -mt_{\mathrm{Co}}H\cos\theta_1 + k_a t_{\mathrm{Co}}\sin^2(\theta_1-\phi) - mt_{\mathrm{Co}}H\cos\theta_2 + k_a t_{\mathrm{Co}}\sin^2(\theta_2-\phi) + 2J_{\mathrm{af}}\cos(\theta_1-\theta_2), \quad (1)$$

where m is the saturation magnetization of Co layers with a thickness t_{Co}, k_a is an anisotropy constant [higher orders of $\sin^2(\theta-\phi)$ are neglected], ϕ is the angle between the easy axis and the x axis, and J_{af} is the AF coupling constant. (We do not include a contribution from biquadratic coupling, which seems to be insignificant in sputtered Co/Cu MLs.[11]) This expression is similar to one given by Folkerts[6] for an AF-coupled bilayer, but differs principally by a factor of 2 in the last term because, with the infinite stack, each Co layer is coupled to two other Co layers, rather than one.

We make use of an AF-order parameter that is defined by

$$AF(H) \equiv \tfrac{1}{2}(1-\cos(\theta_1-\theta_2)), \quad (2)$$

with maximum value AF_{max}. The field-dependent magnetoresistance is defined by

$$MR(H) \equiv \frac{R(H)-R_{\mathrm{sat}}}{R_{\mathrm{sat}}}, \quad (3)$$

with the maximum value MR_{max}, where $R(H)$ is the field-dependent resistance and R_{sat} is the limiting value of the resistance that is attained at large applied magnetic fields. We shall also make use of a normalized magnetoresistance, for which $AF(H)$ is a convenient surrogate,

$$MR^*(H) = \frac{R(H)-R_{\mathrm{sat}}}{R_0-R_{\mathrm{sat}}}, \quad (4)$$

with maximum value MR^*_{max}, where R_0 is the resistance of the as-grown ML. Also, for convenience, we use the shorthand ΔR_{max} and ΔMR_{max} to represent the fractional losses in maximum resistance and maximum magnetoresistance that occur irreversibly with magnetic saturation.

In applying Eq. (1) we make some assumptions about the anisotropy. First, we assume that the crystal anisotropy is not strong enough to force the magnetization vector out of the film plane. Second, we assume uniaxial anisotropy with an easy axis that arises from projection of the closest crystallographic easy axis onto the film plane. Thus, the sine squared function of Eq. (1) is clearly an approximation. This is somewhat justified by our observation that the calculations give similar results with the assumptions of twofold or threefold symmetry, although with different values of the anisotropy constant.

Figure 3 shows energy contours, $E(\theta_1,\theta_2)$, for a Co/Cu ML at the second AFM with four values of H. For illustrative purposes, we have chosen $\phi = 45°$. This is equivalent to one of the cases that was treated algebraically by Folkerts, but a detailed examination of this energy surface facilitates subsequent discussion by illuminating a sequence of states that is qualitatively characteristic of all easy-axis orientations. We assume that the system can become trapped in a local energy minimum, no matter how shallow (i.e., we neglect thermal activation).

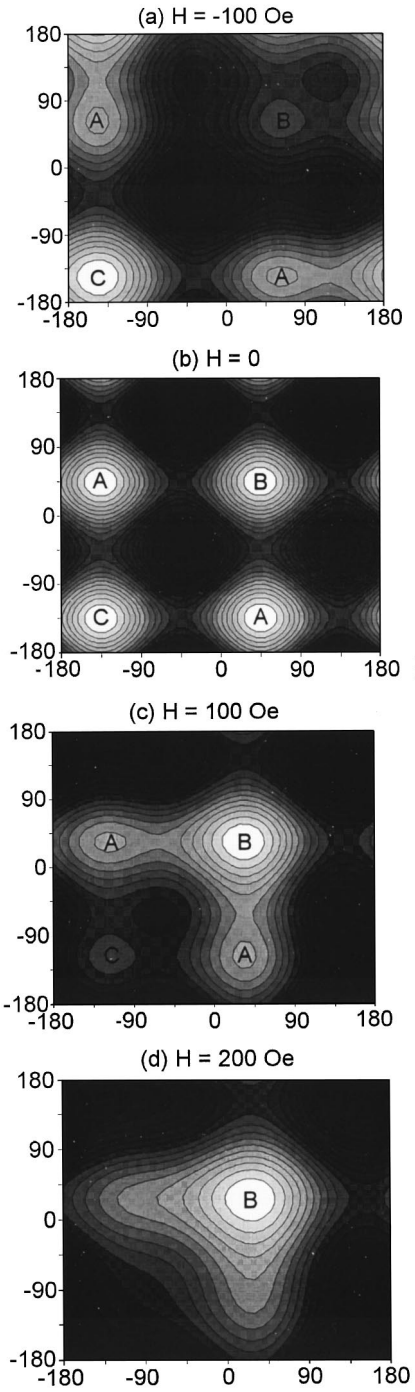

FIG. 3. Energy surfaces for an infinite stack of Co/Cu layers at the second AFM with various values of the applied magnetic field. The coordinates θ_1 and θ_2 are the magnetization angles of alternate Co layers relative to the field direction and lighter shades correspond to lower energies. The calculation takes $m = 1300$ emu/cm^3, $J_{\mathrm{af}} = 0.006$ erg/cm^2, and $k_a = 2\times10^5$ erg/cm^3, with $\phi = 45°$.

Consider first the case with $H = 0$ [Fig. 3(b)]. We postulate that an as-grown ML has the perfect AF order that corresponds to its lowest energy state. Thus, it will occupy one of the two equivalent global minima that are labeled A, in

which alternating Co layers have their magnetization vectors at 45° and 135°, respectively. These correspond simultaneously to AF ordering and to magnetizations along the easy axes. With an increase of the applied field to 100 Oe [Fig. 3(c)] the minima at A shift, but the system that is trapped in them remains approximately AF-ordered. However, at 200 Oe [Fig. 3(d)], the minima at A have disappeared and the system has fallen into the F-ordered minimum at B. Here the two magnetization angles become equal and adopt a field-dependent compromise between $\theta=45°$ that satisfies the anisotropy and $\theta=0$ that satisfies the interaction with the applied field. It is this transition between the minima at A and B that gives the irreversible change that is associated with cycling. If we now decrease the applied field back to zero, the system remains trapped in the minimum at B that persists through $H=0$ [Fig. 3(b)], but disappears by $H\approx-100$ Oe [Fig. 3(a)] when the system has fallen back into the one of the approximately AF-ordered minima at A. With further increase in the reverse field, the minima at A eventually disappear when the system falls into another F-ordered state at C, where the two angles adopt a field-dependent compromise between $\theta=135°$ that satisfies the anisotropy and $\theta=180°$ that satisfies the interaction with the applied field. Hysteresis is manifested in the negative offset from zero applied field of the return to the approximately AF-ordered state after subjection to large enough positive fields.

Note that, when the applied field is swept between extreme values, the system only occupies the minima at A at nonzero values of the field. This implies that there will always be a compromise between satisfying the AF coupling and satisfying the interaction with the magnetic field. For this reason, the ordering is only approximately AF and the resulting resistance after magnetic saturation is always less than that of the as-grown sample. This accounts for much of $\Delta R_{\max}$. (We should emphasize that here the AF ordering is imperfect in the sense that the angles θ_1 and θ_2 do not differ by 180°, rather than because the system has randomness, as was the case with the attempted explanation in terms of ADBs.)

We must also allow for the fact that most Co/Cu MLs (including our own) are made by sputtering and range in structure from randomly-oriented polycrystalline to possession of some degree of fiber texture. (The MLs that were used for the measurements that we describe later were polycrystalline and randomly oriented.) The individual grains of the polycrystalline MLs will then have easy axes that are randomly oriented. This gives rise to a variation in the fields at which the individual grains of a cycled ML pass through the approximately AF-ordered state. This asynchrony in response to the field gives a further decrease in the maximum values of the *average* AF order and hence of the maximum resistance of the polycrystalline ensemble.

Consider a randomly-oriented polycrystalline ML each of whose individual grains maintains epitaxial order from layer to layer. We apply a simple model in which the AF order is averaged over all easy-axis orientations of the grains. Consideration of the end effects due to a finite number of repeat units in the ML is deferred until later. Thermal activation is also neglected. The calculations are made by tracking the movements of the minima of the energy surface that is defined by Eq. (1) as the applied magnetic field is changed by small increments (typically 1 Oe). We use a steepest descent numerical technique with care taken to avoid getting hung up when a local minimum turns into a saddle point. The results are summed over all easy-axis directions with a resolution of 1°. Typical results for an infinite stack of Co/Cu bilayers at the second AFM are shown in Fig. 4, which compares the AF/H curves calculated with zero and increasing anisotropy when the applied field is reversed from large positive values, where the ML has F order. These results show quite clearly a decrease of $AF_{\max}$ as increasing values of the anisotropy constant displace the AF/H peak from zero applied field. This confirms our speculation of a correlation between magnetoresistive hysteresis and $\Delta R_{\max}$.

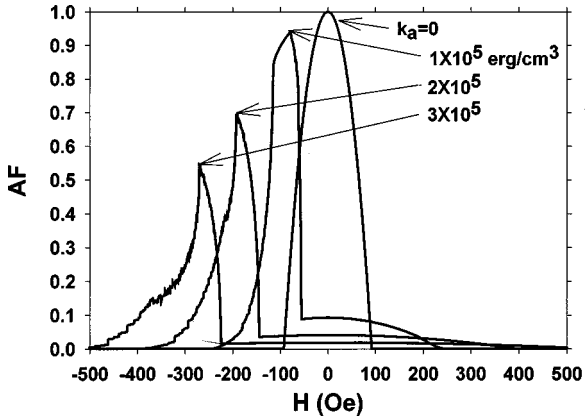

FIG. 4. Influence of the anisotropy constant on the antiferromagnetic order parameter of infinite stacks of Co/Cu layers at the second AFM after cycling. The applied magnetic field is swept from positive to negative values and we take $m=1300$ emu/cm^3, $t_{\mathrm{Co}}=t_{\mathrm{Cu}}=20$ Å, and $J_{\mathrm{af}}=0.006$ erg/cm^2.

We note that in Fig. 4 the curve with $k_a=0$ is close to a parabolic form that would be expected theoretically. For this case in Eq. (1) with approximate AF order we have, by symmetry, $\theta_1=-\theta_2=\theta$ and setting $dE/d\theta=0$ for an infinite ML at equilibrium, we obtain

$$\cos\,\theta=\frac{mt_{\mathrm{Co}}H}{4J_{\mathrm{af}}}, \tag{5}$$

and, since in this case, $\Delta R_{\mathrm{mag}}\propto\frac{1}{2}(1-\cos 2\theta)$,

$$R-R_{\mathrm{sat}}\propto 1-\left(\frac{mt_{\mathrm{Co}}H}{4J_{\mathrm{af}}}\right)^2. \tag{6}$$

At saturation $\theta=0$ and we obtain a result that has been derived previously,[12] i.e.,

$$J_{\mathrm{af}}=\frac{mt_{\mathrm{Co}}H_{\mathrm{sat}}}{4}. \tag{7}$$

Taking $m=1300$ emu/cm^2, for fcc Co, $J_{\mathrm{af}}=0.006$ erg/cm^2, and $t_{\mathrm{Co}}=20$ Å, the curve obtained by tracking the energy minima gives $AF\rightarrow 0$ at $H=92$ to 93 Oe in excellent agreement with the exact calculation from Eq. (7), which gives $H_{\mathrm{sat}}=92.3$ Oe. This provides a useful verification of our numerical minimum-tracking procedure.

Figure 4 illustrates an important point that bears on the usual derivation of the coupling constant from the saturation

field via Eq. (7). When anisotropy is introduced, the AF/H peak at first narrows and similar behavior is to be expected from the R/H peak. This can introduce an error into the value that is deduced for the coupling constant because Eq. (7) is no longer valid when anisotropy is significant.

We have yet to justify our use of the average AF order parameter as a surrogate for the magnetoresistance. It has already been established theoretically and experimentally that this is appropriate for a singly-oriented ML[13,14] and there is experimental evidence that a similar relationship holds for polycrystalline sputtered MLs.[15–17] The problem that remains is to show why this is a reasonable approximation for a polycrystalline ML when the differently oriented grains are at different stages in the transition between maximum and minimum resistance. We note that the resistivity of the polycrystalline ML must be bounded by two extreme values. These correspond to the combination of the individual grain resistivities in series and in parallel, respectively. Of these two cases, the series combination corresponds exactly to taking the arithmetic average of the resistivities and hence of the AF order. It is easy to show that the alternative parallel combination will differ from the series combination only by terms of second or higher order in the resistances.

For a more formal approach to the same conclusion, we note that Rossiter[18] gives a result for the conductivity, σ^*, of a random assembly of cylinders with conductivities σ_1, σ_2 and volume fractions ν_1, ν_2 when the current is perpendicular to the cylinder axes. This is a reasonable description of the in-plane conductivity of a polycrystalline sputtered film, which would be expected to exhibit a columnar growth habit. Rossiter's result is then

$$\nu_1\left(\frac{\sigma_1-\sigma^*}{\sigma_1+\sigma^*}\right)+\nu_2\left(\frac{\sigma_2-\sigma^*}{\sigma_2+\sigma^*}\right)=0, \tag{8}$$

which is expected to be a fair approximation when the conductivities of the components do not differ greatly. For the special case when the volume fractions are equal this gives the result that the resulting resistivity is simply the geometric mean of that of the components, i.e., $\rho^*=\sqrt{\rho_1\rho_2}$. By induction, this may be extended to our case with a large number of randomly arranged grain orientations that are present in equal volumes. Simply replace component 1 with an equal mixture of components 3 and 4 and replace component 2 with an equal mixture of components 5 and 6. Repetition of this process and substitution into the definition of magnetoresistance gives

$$(1+MR^*)^n=(1+MR_1)(1+MR_2)\cdots(1+MR_n), \tag{9}$$

whence, to first order in the magnetoresistances,

$$MR^*=\overline{MR}=MR_{\max}\overline{\mathrm{AF}}. \tag{10}$$

IV. REFINEMENT OF THE MODEL FOR A FINITE STACK

Now we consider the consequences of having a finite, rather than an infinite, stack of bilayers. Clearly, the outermost ferromagnetic layers differ by being AF-coupled on only one side and their magnetization angles will reflect this. Moreover, the perturbation of the outer layers of the finite stack that is introduced by its termination will propagate to the neighboring layers. Such a situation has been considered by Mattheis *et al.*[19] but only for a case in which there is no in-plane anisotropy. In the general case with in-plane anisotropy and a finite ML with N bilayers, we must modify Eq. (1) to obtain a total energy per unit area

$$E=-\sum_{j=1}^{N} mt_{\mathrm{Co}}H\cos\theta_j+\sum_{j=1}^{N} k_a t_{\mathrm{Co}}\sin^2(\theta_j-\phi)+\sum_{j=1}^{N-1} J_{\mathrm{af}}\cos(\theta_j-\theta_{j+1}). \tag{11}$$

Solution for the local minima of this N-dimensional energy surface is then effected by steepest descents in the same way as for the two-dimensional case. Once more we need to sum over all easy-axis orientations of a polycrystalline assembly of grains. We must also extend our definition of the AF order parameter to average over all pairs of adjacent Co layers.

$$AF\equiv\frac{1}{N-1}\sum_{j=1}^{N-1}\tfrac{1}{2}[1-\cos(\theta_j-\theta_{j+1})]. \tag{12}$$

An example of the influence of finite size is shown in Fig. 5. In general, the peak in the AF/H curve is reduced in height and this effect remains substantial with as many as 20 bilayers. The further reduction in $AF_{\max}$ that occurs here is a consequence of the reduction of the average strength of the AF coupling due to the end effects. This enhances the influence of anisotropy.

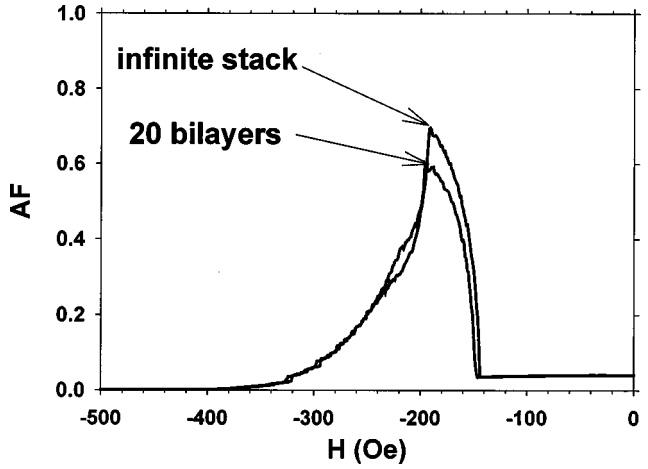

FIG. 5. Influence of a finite (20 bilayer) Co/Cu stack on the AF order parameter when the field is swept from positive to negative values after cycling. We take $m=1300$ emu/cm^3, $t_{\mathrm{Co}}=t_{\mathrm{Cu}}=20$ Å, and $J_{\mathrm{af}}=0.006$ erg/cm^2.

Figure 6 shows the effect of increasing anisotropy on the height and position of the AF peak for finite, 20 bilayer, Co/Cu MLs at the second AFM. The results follow the same trend as for infinite stacks, but with somewhat reduced peak heights. For these calculations and those that follow we have taken a value of the saturation magnetization for fcc Co, rather than the hcp phase that constitutes the bulk material at 300 K. Magnetic resonance measurements[20,21] have shown that the fcc Co phase is the major component in our multilayers. We have chosen the value $m=1300$ emu/cm^3 from a

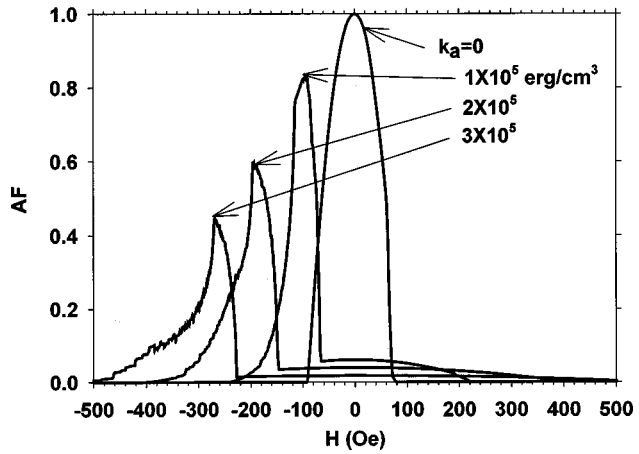

FIG. 6. Influence of the anisotropy constant on the antiferromagnetic order parameter of 20-bilayer stacks of Co/Cu at the second AFM after cycling. The applied magnetic field is swept from positive to negative values and we take $m=1300$ emu/cm^3, $t_{\mathrm{Co}}=t_{\mathrm{Cu}}=20$ Å, and $J_{\mathrm{af}}=0.006$ erg/cm^2.

published survey of the literature,[22] although there is significant uncertainty and the experimental work in Ref. 22 suggests a somewhat smaller value. However, the difference from the value for hcp Co ($m\approx 1400$ emu/cm^3 in the temperature range of interest) does not greatly affect the results.

Figure 7 shows that, with a particular value of the anisotropy constant, the peak in the AF/H curve decreases with decrease of the AF coupling constant while its position remains approximately constant. The influence of Co thickness on the peak height is considered later in Sec. VI.

V. APPLICATION OF THE MODEL TO Co/Cu MLs

We continue by using our model to discuss the experimentally determined properties of Co/Cu MLs. These were grown by dc magnetron sputtering onto oxidized Si substrates with 75 Å Ru buffer layers using techniques that have been fully described elsewhere.[8–10] Electron diffraction patterns of typical MLs, taken in transmission, showed no arcing on tilting, which indicated an absence of significant texturing.[23]

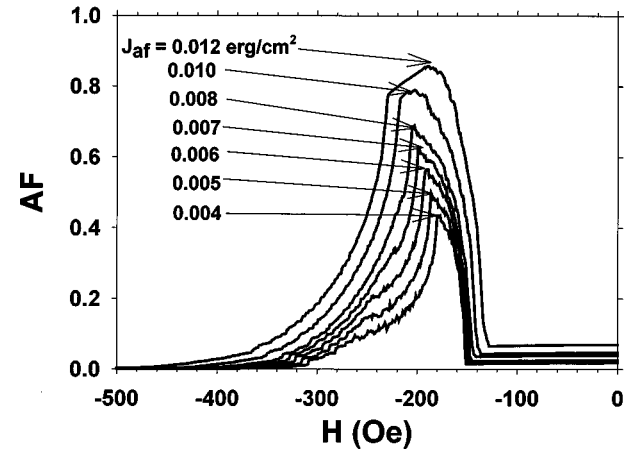

FIG. 7. Influence of the antiferromagnetic coupling constant on the antiferromagnetic order parameter of 20-bilayer stacks of Co/Cu at the second AFM after cycling. The applied magnetic field is swept from positive to negative values and we take $m=1300$ emu/cm^3, $t_{\mathrm{Co}}=t_{\mathrm{Cu}}=20$ Å, and $k_a=2\times 10^5$ erg/cm^3.

For convenience, we summarize the postulates of the model as follows:

(i) The grains of an as-grown polycrystalline ML have perfect AF order, corresponding to the state of lowest energy.
(ii) The polycrystalline grains have in-plane magnetic anisotropy.
(iii) The system can become trapped in local energy minima. (This assumes that thermal activation is negligible.)
(iv) We include the end effects that occur with finite ML stacks. (Typically comprised of 20 Co/Cu bilayers.)
(v) Intergrain magnetostatic interactions are neglected.

We shall treat the anisotropy constant and the antiferromagnetic coupling constant as adjustable parameters and subsequently consider the reasonableness of the values that we obtain.

VI. Co/Cu MLs AT THE SECOND AFM

Our Co/Cu MLs exhibit a significant decrease in $MR_{\max}$ when t_{Co} is made larger than about 15 Å and we have been informed of a similar result elsewhere.[24] Also, Shukh *et al.*[25] have reported decreasing $MR_{\max}$ for t_{Co} larger than 10 Å. Such results are quite strikingly different from those at the first AFM[26] where $MR_{\max}$ decreases only slowly and approximately linearly as t_{Co} is increased in the range 5–50 Å. As we shall demonstrate, the decrease in $MR_{\max}$ with increasing t_{Co} at the second AFM is mostly due to an increase in $\Delta R_{\max}$.

From Eqs. (1) and (11) it is evident that the effects of anisotropy are enhanced by increase of $k_a t_{\mathrm{Co}}/J_{\mathrm{af}}$. On going from the first to the second AFM, J_{af} decreases approximately fourfold. This is consistent with the observed increases at the second AFM of magnetoresistive hysteresis and of $\Delta R_{\max}$ relative to the first AFM. From this one might expect that, once hysteresis becomes significant, increasing t_{Co} would give larger values of $\Delta R_{\max}$, provided that k_a does not vary greatly with t_{Co}. This would be expected to contribute to the observed drop in $MR_{\max}$. The experimental data in Fig. 8 show that this is indeed the case. $\Delta MR_{\max}$ at 300 K increases from $\sim$10% at $t_{\mathrm{Co}}=10$ Å to $\sim$50% at $t_{\mathrm{Co}}=20$ Å, while the as-grown value of $MR_{\max}$ remains in the range 21%–26%. (If spin-dependent scattering occurs mostly at the Co/Cu interfaces,[27,28] the magnetoresistance of the as-grown samples would be expected to decrease only linearly with the sum of the Co and Cu thicknesses.) These results are typical of many specimens that give a usual range of variation of only a few % in $MR_{\max}$ as grown.

Figure 9 compares typical experimental values of $MR^*(H)$ for Co/Cu MLs at the second AFM with calculated values of $AF(H)$. In choosing the parameters for the AF/H curves we have first selected a value of k_a to give approximately the correct peak offset from zero applied field and then adjusted J_{af} to approximately fit the peak heights.

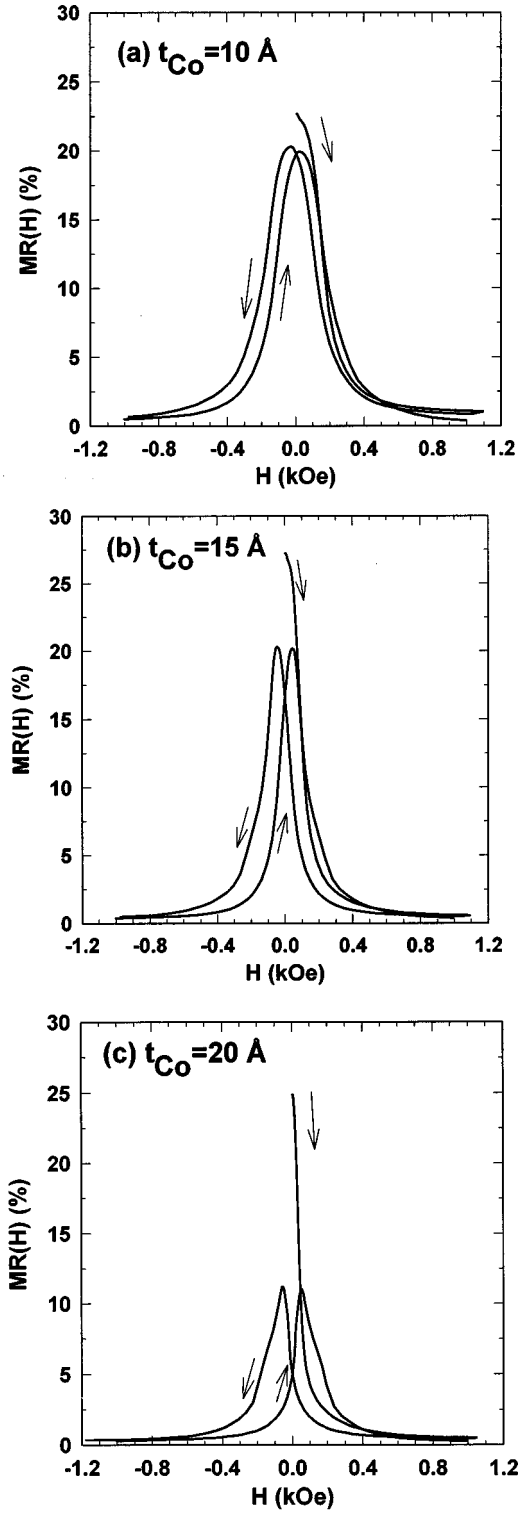

FIG. 8. Experimental 300 K magnetoresistance curves as grown and after cycling of Co/Cu multilayers at the second AFM. The structures are $[\mathrm{Co}(t_{\mathrm{Co}})/\mathrm{Cu}20\ \text{Å})]_{20}$ with (a) $t_{\mathrm{Co}}=10$ Å. (b) $t_{\mathrm{Co}}=15$ Å. (c) $t_{\mathrm{Co}}=20$ Å.

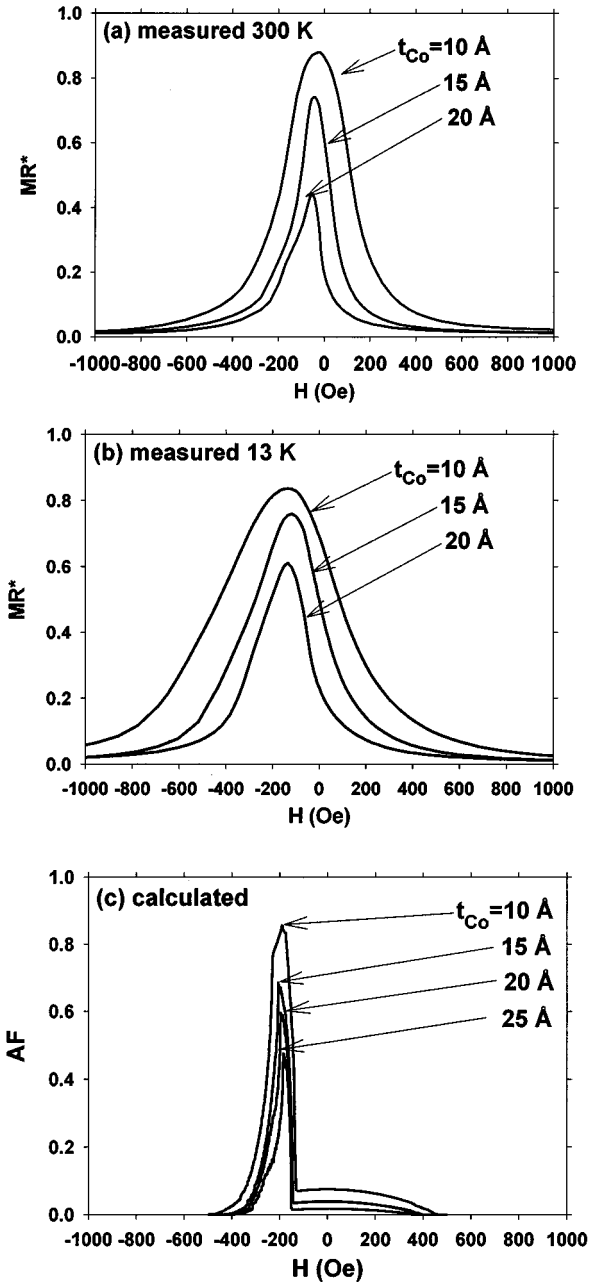

FIG. 9. Experimental values of the MR^*_{max} at the second AFM after cycling compared with calculated values of the maximum antiferromagnetic order parameter. The experimental structures are $[\mathrm{Co}(t_{\mathrm{Co}})/\mathrm{Cu}20\ \text{Å})]_{20}$ with $t_{\mathrm{Co}}=10$, 15, and 20 Å. The applied magnetic field is swept from positive to negative values and the calculation takes $m=1300$ emu/cm^3, $J_{\mathrm{af}}=0.006$ erg/cm^2, and $k_a=2\times10^5$ erg/cm^3. (a) Measured at 300 K. (b) Measured at 13 K. (c) Calculated.

The agreement of the calculated $AF(H)$ with the peak heights and positions of the experimental $MR^*(H)$ is reasonable, both at 300 K and at 13 K, although the calculated peaks are substantially narrower than is observed experimentally. A comparison of the calculated AF_{max} and measured MR^*_{max} for a wider range of experimental data is given in Fig. 10. There is some scatter in the experimental data, but overall the agreement with the calculated dependence on t_{Co} is excellent both at 300 K and at 13 K. From this we can conclude that the anisotropy constant is not strongly dependent on t_{Co}. Also, the insensitivity of the experimental results to temperature provides strong justification for our neglect of thermal activation.

VII. BEHAVIOR AT OTHER AFMS

As discussed earlier in this article, we would expect ΔR_{max} to be a function of $k_a t_{\mathrm{Co}}/J_{\mathrm{af}}$. Thus, for k_a not being

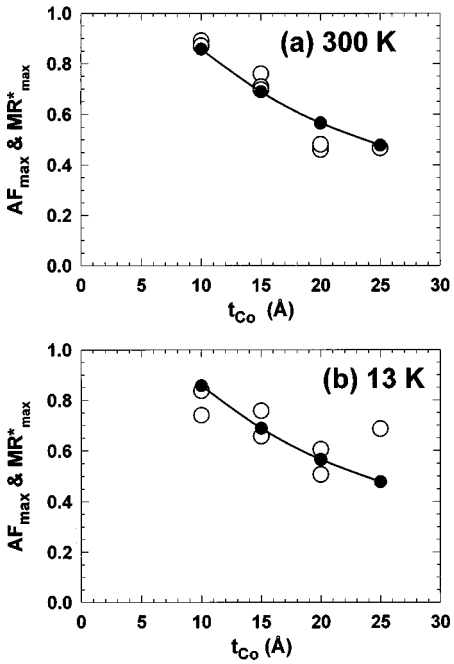

FIG. 10. Comparisons of the calculated values of the maximum value of the antiferromagnetic order parameter after cycling with experimental values of $MR^*_{\max}$ as a function of t_{Co}. (a) Measurements at 300 K. (b) Measurements at 13 K. In both (a) and (b), the calculations are identical with parameters $m=1300$ emu/cm^3, $t_{\mathrm{Cu}}=20$ Å, $J_{\mathrm{af}}=0.006$ erg/cm^2, and $k_a=2\times10^5$ erg/cm^3.

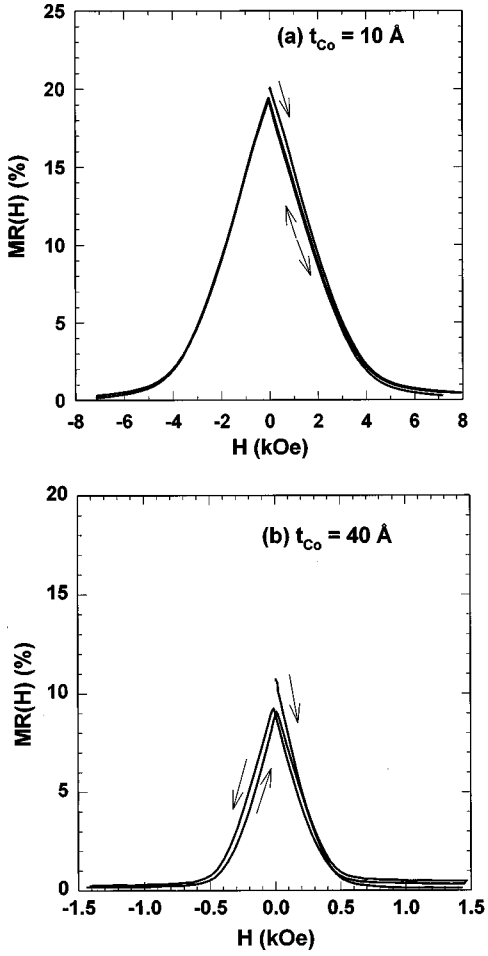

FIG. 11. Experimental 300 K magnetoresistance curves as grown and after cycling of Co/Cu multilayers at the first AFM. The structures are $[\mathrm{Co}(t_{\mathrm{Co}})/\mathrm{Cu}9\ \text{Å})]_{20}$. (a) $t_{\mathrm{Co}}=10$ Å. (b) $t_{\mathrm{Co}}=40$ Å.

a strong function of t_{Co} we should expect that, at the first AFM, the resistance loss on cycling would become significant with values of t_{Co} about four times those required for such an effect at the second AFM. Figure 11 shows that this is, indeed, the case; while $\Delta R_{\max}$ is small when $t_{\mathrm{Co}}=10$ Å, it increases to about 10% when $t_{\mathrm{Co}}=40$ Å.

At AFMs we expect[29] to have $J_{\mathrm{af}}\propto 1/t_{\mathrm{Cu}}^2$, whence the argument above leads to a prediction that $MR^*_{\max}$ will be a function of $t_{\mathrm{Co}}t_{\mathrm{Cu}}^2$ that will apply irrespective of the choice of AFM. Figure 12 compares our calculations of $AF_{\max}$ with experimental values of $MR^*_{\max}$ for the first through fourth AFMs at 300 and 13 K. (The calculations take $J_{\mathrm{af}}=0.006$ erg/cm^2 at the second AFM and $J_{\mathrm{af}}\propto 1/t_{\mathrm{Cu}}^2$ at other AFMs.) As with the second AFM, the experimental data for all other AFMs are essentially independent of temperature, which further validates our neglect of thermal activation. We see that the first and second AFMs agree quite well with the calculation, but that the third and fourth AFMs deviate significantly giving values of $MR^*_{\max}$ that are larger than calculated. It seems likely that this deviation arises from a breakdown of our postulate that the as-grown MLs have perfect AF order. Published curves of the dependence of $MR_{\max}$ on t_{Cu} in Co/Cu MLs show that $MR_{\max}$ is approximately zero between the first and second AFMs ($t_{\mathrm{Cu}}\approx 14$ Å) where F coupling is expected, but that zero magnetoresistance is not obtained between the second and third or between the third and fourth AFMs where again F coupling is expected. (See Ref. 30, Fig. 3 and Ref. 31, Fig. 2.) This implies that the F coupling at these large Cu separator thicknesses is not strong enough to impose complete F order. One might then expect comparably weak AF coupling to lead also to incomplete AF order in the same range of Cu thicknesses. This would give rise to larger $MR^*_{\max}$ because the MLs start with less AF order.

VIII. DISCUSSION

The model that we have presented provides an explanation for the irreversible decrease in maximum resistance that occurs after magnetoresistive MLs are magnetically saturated and it accounts quantitatively for the relatively small effect at the first AFM and the dependence on t_{Co} at the second AFM. We must now consider the reasonableness of the two parameters that we have used to fit this wide range of data. First, our value $k_a=2\times10^5$ erg/cm^3 is not too different from an experimental value for fcc Co films[22] of 5×10^5 erg/cm^2 and the agreement here may be regarded as adequate considering the likely variation of this parameter with film orientation and substrate-induced strain.

Our fitted value of the AF coupling constant, $J_{\mathrm{af}}=0.006$ erg/cm^2 at the second AFM, is somewhat smaller than expected. An experimental value derived from the saturation fields of coupled bilayers[29] (and subject to the reservation about validity that is discussed in Sec. III) is $J_{\mathrm{af}}\approx 0.05$ erg/cm^2. However, this was obtained using a relation

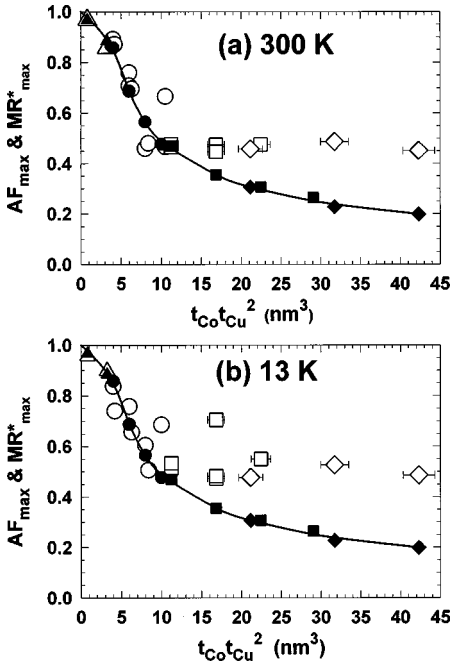

FIG. 12. Comparisons of the calculated values of the maximum value of the antiferromagnetic order parameter after cycling with experimental values of $MR^*_{\max}$ as a function of $t_{\mathrm{Co}}t_{\mathrm{Cu}}^2$. The filled symbols are calculated points and the open symbols are experimental data. ▲,△ first AFM. ●,○ second AFM. ■,□ third AFM. ♦,◇ fourth AFM. (a) Measurements at 300 K. (b) Measurements at 13 K. In both (a) and (b), the calculations are identical and take $m=1300$ emu/cm^3 and $k_a=2\times10^5$ erg/cm^3. J_{af} is scaled to be $\propto 1/t_{\mathrm{Cu}}^2$, i.e., with values 0.0296, 0.006, 0.00214, and 0.00113 erg/cm^2 for the 1st through 4th AFMs corresponding to $t_{\mathrm{Cu}}=9$, 20, 33.5, and 46 Å. The error bars are based on assumed uncertainties of ± 0.5 Å in each of t_{Co} and t_{Cu} and the line is a slightly smoothed fit to the calculations.

for a bilayer (transcribed to the present notation) $J_{\mathrm{af}}=mt_{\mathrm{Co}}H_{\mathrm{sat}}$ which is overlarge by a factor of two.[12] (The correct result may be derived easily with minor modification of the argument given for an infinite stack in Sec. III above.) Even after this correction, the published experimental value is four times larger than our fitting parameter. It would not be surprising were J_{af} to vary to some extent with the orientation and the interfacial roughness of the MLs, which in turn will depend upon the growth conditions. Some evidence for such a variation in J_{af} is provided by a variation in the value of t_{Co} at which the magnetoresistance of second AFM specimens suffers substantial loss after cycling. Thus, a cycled (111)-textured specimen with $t_{\mathrm{Co}}=20$ Å grown at Michigan State University[32] gave $MR_{\max}=24.6\%$ at 300 K, which is much larger than we obtain at this thickness [see Fig. 8(c)] and would imply stronger coupling. In contrast, cycled specimens grown at the Demokritos Institute, Athens[24] show a substantial decrease in magnetoresistance with $t_{\mathrm{Co}}>10$ Å, which would imply weaker coupling.

Despite the likelihood of some variation in AF coupling strength with variation of the growth process, we still suspect that our fitted value of J_{af} is too small, perhaps by a factor of two. This tentative conclusion is drawn from the fact that our AF/H peaks are substantially narrower than the experimental R/H peaks. This is the case not only at the second AFM, but also at the first AFM where the width is negligibly affected by anisotropy.[33]

We speculate that our fitting parameters may be significantly influenced by our neglect of intergrain magnetostatic interactions. This is a problem that, to date, we have found intractable. In a simple effective-medium approach each grain in the polycrystalline assembly will be subject to an effective field

$$H_{\mathrm{eff}}=H_{\mathrm{appl}}+A(M_{\mathrm{av}}-M_{\mathrm{loc}}), \tag{13}$$

where H_{appl} is the applied magnetic field, M_{av} is the average magnetization of the surrounding medium, M_{loc} is the local magnetization of the grain, and A is a constant that depends on the grain shape. (For a spherical grain $A=4\pi/3$ and for a cylindrical grain that would approximate the columnar texture of sputtered films $A=2\pi$.) It would appear to be a simple matter to apply Eq. (13) iteratively to the polycrystalline assembly, but one runs into problems with convergence of the result. Further work on this problem is in progress.

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[33] With a more realistic model for the anisotropy we should consider a range of effective anisotropy constants that arises from randomness of the inclination of the nearest crystallographic easy axis to the film plane. We should then sum over values of $k_a = k_a' \cos^2 \psi$, where k_a' is the crystallographic anisotropy constant and ψ is the angle between the easy axis and the plane. This would tend to broaden the AF/H peaks at the second AFM, but not at the first AFM, where the anisotropy is relatively insignificant.